

Application No.: H/596,465 10/570,025

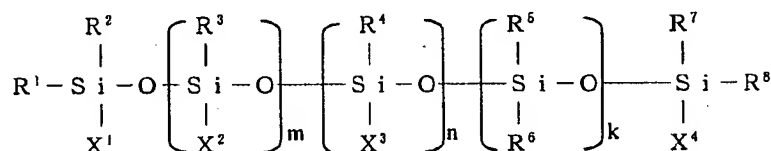
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AMENDED CLAIM SET:

1. (currently amended) A conjugated diene rubber composition comprising:

(A) 5% to 95% by weight of a conjugated diene rubber having a weight average molecular weight in the range of 1,000 to 3,000,000 wherein at least three conjugated diene polymer chains in said rubber are bonded together through at least one polyorganosiloxane selected from those which are represented by the general formulae (1), (2) and (3), shown below:

General formula (1):



wherein

R¹ through R⁸ represent an alkyl group having 1 to 6 carbon atoms or an aryl group having 6 to 12 carbon atoms, R¹ through R⁸ being the same or different;

~~X¹ and X⁴ are such that~~ (i) a part of the sum of X¹ and X⁴ is a functional group capable of reacting with an active metal at a terminal of the active conjugated diene polymer chains selected from the group consisting of a hydrocarbon group containing a 2-pyrrolidonyl group and an epoxy-containing group having 4 to 12 carbon atoms and the remainder of the sum of X¹ and X⁴ is a group derived from said functional group by the reaction of said functional group with an active metal at a terminal of the active conjugated diene polymer chains or a single bond, or (ii) a part of the sum of X¹ and X⁴ is an alkoxyl group having 1 to 5 carbon atoms and the remainder of the sum of X¹ and X⁴ is a single bond, or (iii) X¹ and X⁴ are an alkyl group having 1 to 6 carbon atoms or an aryl group having 6 to 12 carbon atoms, X¹ and X⁴ being the same or different;

~~X² is such that~~ (i) a part of X² is a functional group selected from the group consisting of a hydrocarbon group containing a 2-pyrrolidonyl group and an epoxy-containing group having 4 to 12 carbon atoms capable of reacting with an active metal at a terminal of the active conjugated diene polymer chains and the remainder of X² is a group derived from said functional group by

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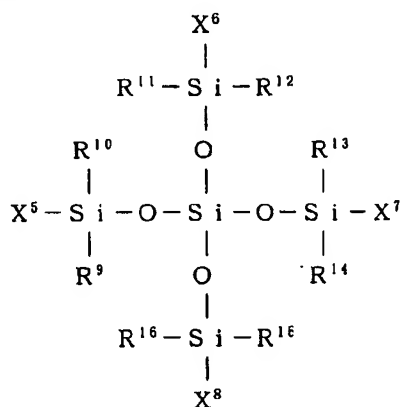
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the reaction of said functional group with an active metal at a terminal of the active conjugated diene polymer chains, or (ii) a part of X^2 is an alkoxyl group having 1 to 5 carbon atoms and the remainder of X^2 is [[or]] a single bond;

X^3 is a group comprising 2 to 20 alkylene glycol repeating units, provided that a part of X^3 may be a group derived from the group comprising 2 to 20 alkylene glycol repeating units; and

m is an integer of 3 to 200, n is an integer of 0 to 200 and k is an integer of 0 to 200;

General formula (2):

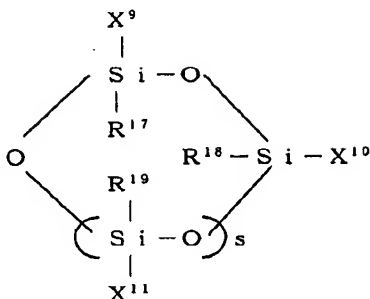


wherein

R^9 through R^{16} represent an alkyl group having 1 to 6 carbon atoms or an aryl group having 6 to 12 carbon atoms, R^9 through R^{16} being the same or different; and

~~X^5 through X^8 are such that~~ (i) a part of the sum of X^5 through X^8 is a functional group selected from the group consisting of a hydrocarbon group containing a 2-pyrrolidonyl group and an epoxy-containing group having 4 to 12 carbon atoms capable of reacting with an active metal at a terminal of the active conjugated diene polymer chains and the remainder of the sum of X^5 through X^8 is a group derived from said functional group by the reaction of said functional group with an active metal at a terminal of the active conjugated diene polymer chains, or (ii) a part of the sum of X^5 through X^8 is an alkoxyl group having 1 to 5 carbon atoms and the remainder of the sum of X^5 through X^8 is [[or]] a single bond;

General formula (3):



wherein

R¹⁷, R¹⁸ and R¹⁹ represent an alkyl group having 1 to 6 carbon atoms or an aryl group having 6 to 12 carbon atoms, R¹⁷, R¹⁸ and R¹⁹ being the same or different;

X^9 , X^{10} and X^{11} are such that (i) a part of the sum of X^9 , X^{10} and X^{11} is a functional group selected from the group consisting of a hydrocarbon group containing a 2-pyrrolidonyl group and an epoxy-containing group having 4 to 12 carbon atoms capable of reacting with an active metal at a terminal of the active conjugated diene polymer chains and the remainder of the sum of X^9 , X^{10} and X^{11} X^1 and X^4 is a group derived from said functional group by the reaction of said functional group with an active metal at a terminal of the active conjugated diene polymer chains, or (ii) a part of the sum of X^9 , X^{10} and X^{11} is an alkoxy group having 1 to 5 carbon atoms and the remainder of the sum of X^9 , X^{10} and X^{11} is $[(\text{or})]$ a single bond; and

s is an integer of 1 to 18, and

(B) 95% to 5% by weight of ~~a reaction product of~~ a conjugated diene rubber having a weight average molecular weight in the range of 1,000 to 3,000,000 which is a reaction product of an active conjugated diene polymer chain having an active metal at a terminal thereof with a compound selected from the group consisting of:

- N-substituted cyclic amides,
N-substituted cyclic ureas,
N-substituted amino ketones,
aromatic isocyanates,
N,N,N',N'-tetramethylthiourea,

N,N-disubstituted aminoalkylmethacrylamides,
N-substituted aminoaldehydes,
N-substituted carbodiimides,
Schiff bases,
propylene oxide,
tetraglycidyl-1,3-bisaminomethylcyclohexane,
epoxidized polybutadiene,
vinyl compounds having a pyridyl group,
bis(triethoxysilylpropyl)-tetrasulfide,
bis(tributoxysilylpropyl)tetrasulfide,
 γ -glydoxypropyltrimethoxysilane,
~~methyltrimethoxysilane,~~
tetramethoxysilane,
tin tetrachloride,
silicon tetrachloride,
triphenylmonochlorotin,
triphenoxychlorosilane,
methyltriphenoxysilane, and
diphenoxydichlorosilane.

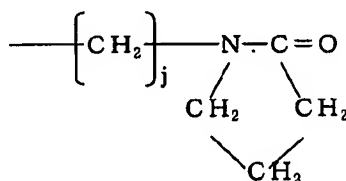
2. (original) The conjugated diene rubber composition according to claim 1, wherein the polymer chain constituting the conjugated diene rubber (A) and the polymer chain constituting the conjugated diene rubber (B) comprise 50% to 100% by weight of conjugated diene monomer units and 50% to 0% by weight of aromatic vinyl monomer units.

3. (original) The conjugated diene rubber composition according to claim 1, wherein the conjugated diene monomer units in the conjugated diene rubber (A) and the conjugated diene monomer units in the conjugated diene rubber (B) have a vinyl bond content of 5% to 95% by weight.

4. (cancelled).

5. (currently amended) The conjugated diene rubber composition according to claim 1 [[4]], wherein the polyorganosiloxane is represented by the general formula (1), and the functional group capable of reacting with an active metal at a terminal of the active conjugated diene polymer chains for X^1 , X^2 and X^4 in the formula (1) is a hydrocarbon group containing a 2-pyrrolidonyl group represented by the following general formula (4).

General formula (4):



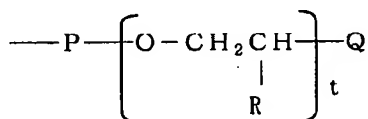
wherein j is an integer of 2 to 10.

6. (currently amended) The conjugated diene rubber composition according to claim 1 [[4]], wherein the polyorganosiloxane is represented by the general formula (1), and the functional group capable of reacting with an active metal at a terminal of the active conjugated diene polymer chains for X^1 , X^2 and X^4 in the formula (1) is a group having 4 to 12 carbon atoms and containing an epoxy group represented by the following general formula (5):

Z Y E

wherein Z is an alkylene group or an alkylarylene group, which have 1 to 10 carbon atoms, Y is a methylene group, a sulfur atom or an oxygen atom, and E is a group having 4 to 12 2 to 10 carbon atoms and containing an epoxy group.

7. (original) The conjugated diene rubber composition according to claim 1, wherein the group comprising 2 to 20 alkylene glycol repeating units for X^3 in the formula (1) is a group represented by the following general formula (6):



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wherein t is an integer of 2 to 20, P is an alkylene group or an alkylarylene group, which have 2 to 10 carbon atoms, R is a hydrogen atom or a methyl group, Q is an alkoxyl group or an aryloxy group, which have 1 to 10 carbon atoms, provided that a part of Q may be a single bond.

8. (currently amended) The conjugated diene rubber composition according to claim 1, wherein the conjugated diene rubber (A) comprises 2 to 90% by weight, based on the conjugated diene rubber (A), of a conjugated diene rubber ~~having a structure such that~~ wherein at least four of the conjugated diene polymer chains in said rubber are bonded together through at least one polyorganosiloxane selected from those which are represented by the formulae (1), (2) and (3).

9. (original) The conjugated diene rubber composition according to claim 1, which further comprises 5 to 150 parts by weight, based on 100 parts by weight of the total conjugated diene rubbers (A) and (B), of at least one filler selected from silica and carbon black.

10. (original) The conjugated diene rubber composition according to claim 9, which comprises silica alone or both of silica and carbon black as the filler.

11. (original) The conjugated diene rubber composition according to claim 1, which further comprises not larger than 900 parts by weight, based on 100 parts by weight of the total conjugated diene rubbers (A) and (B), of a polymer having a glass transition temperature in the range of -120°C to 200°C and a weight average molecular weight of 1,000 to 3,000,000.

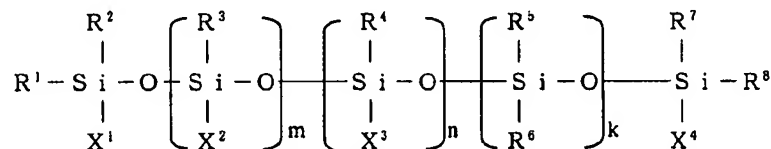
12. - 16. (cancelled).

17. (currently amended) A rubber vulcanizate which is obtained by crosslinking a conjugated diene rubber composition comprising:

(A) 5% to 95% by weight of a conjugated diene rubber having a weight average molecular weight in the range of 1,000 to 3,000,000 wherein at least three conjugated diene

polymer chains in said rubber are bonded together through at least one polyorganosiloxane selected from those which are represented by the general formulae (1), (2) and (3), shown below:

General formula (1):



wherein

R¹ through R⁸ represent an alkyl group having 1 to 6 carbon atoms or an aryl group having 6 to 12 carbon atoms, R¹ through R⁸ being the same or different;

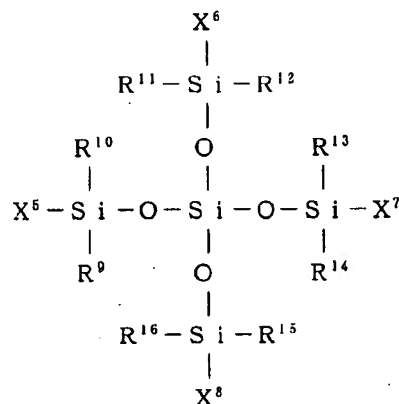
~~X¹ and X⁴ are such that~~ (i) a part of the sum of X¹ and X⁴ is a functional group ~~capable of reacting with an active metal at a terminal of the active conjugated diene polymer chains selected from the group consisting of a hydrocarbon group containing a 2-pyrrolidonyl group and an epoxy-containing group having 4 to 12 carbon atoms and the remainder of the sum of X¹ and X⁴ is a group derived from said functional group by the reaction of said functional group with an active metal at a terminal of the active conjugated diene polymer chains or a single bond~~, or (ii) a part of the sum of X¹ and X⁴ is an alkoxyl group having 1 to 5 carbon atoms and the remainder of the sum of X¹ and X⁴ is a single bond, or (iii) X¹ and X⁴ are an alkyl group having 1 to 6 carbon atoms or an aryl group having 6 to 12 carbon atoms, X¹ and X⁴ being the same or different;

~~X² is such that~~ (i) a part of X² is a functional group selected from the group consisting of a hydrocarbon group containing a 2-pyrrolidonyl group and an epoxy-containing group having 4 to 12 carbon atoms capable of reacting with an active metal at a terminal of the active conjugated diene polymer chains and the remainder of X² is a group derived from said functional group by the reaction of said functional group with an active metal at a terminal of the active conjugated diene polymer chains, or (ii) a part of X² is an alkoxyl group having 1 to 5 carbon atoms and the remainder of X² is [[or]] a single bond;

X^3 is a group comprising 2 to 20 alkylene glycol repeating units, provided that a part of X^3 may be a group derived from the group comprising 2 to 20 alkylene glycol repeating units; and

m is an integer of 3 to 200, n is an integer of 0 to 200 and k is an integer of 0 to 200;

General formula (2):

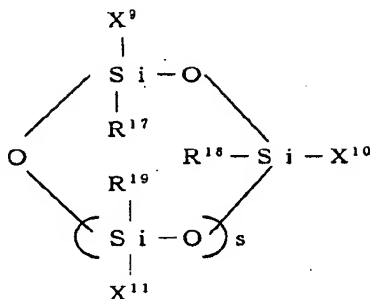


wherein

R^9 through R^{16} represent an alkyl group having 1 to 6 carbon atoms or an aryl group having 6 to 12 carbon atoms, R^9 through R^{16} being the same or different; and

X^5 through X^8 are such that (i) a part of the sum of X^5 through X^8 is a functional group selected from the group consisting of a hydrocarbon group containing a 2-pyrrolidonyl group and an epoxy-containing group having 4 to 12 carbon atoms capable of reacting with an active metal at a terminal of the active conjugated diene polymer chains and the remainder of the sum of X^5 through X^8 is a group derived from said functional group by the reaction of said functional group with an active metal at a terminal of the active conjugated diene polymer chains, or (ii) a part of the sum of X^5 through X^8 is an alkoxyl group having 1 to 5 carbon atoms and the remainder of the sum of X^5 through X^8 is [[or]] a single bond;

General formula (3):



wherein

R¹⁷, R¹⁸ and R¹⁹ represent an alkyl group having 1 to 6 carbon atoms or an aryl group having 6 to 12 carbon atoms, R¹⁷, R¹⁸ and R¹⁹ being the same or different;

X^9, X^{10} and X^{11} are such that (i) a part of the sum of X^9, X^{10} and X^{11} is a functional group selected from the group consisting of a hydrocarbon group containing a 2-pyrrolidonyl group and an epoxy-containing group having 4 to 12 carbon atoms capable of reacting with an active metal at a terminal of the active conjugated diene polymer chains and the remainder of the sum of X^9, X^{10} and X^{11} X^1 and X^4 is a group derived from said functional group by the reaction of said functional group with an active metal at a terminal of the active conjugated diene polymer chains, or (ii) a part of the sum of X^9, X^{10} and X^{11} is an alkoxyl group having 1 to 5 carbon atoms and the remainder of the sum of X^9, X^{10} and X^{11} is [[or]] a single bond; and

s is an integer of 1 to 18, and

(B) 95% to 5% by weight of ~~a reaction product of~~ a conjugated diene rubber having a weight average molecular weight in the range of 1,000 to 3,000,000 which is a reaction product of an active conjugated diene polymer chain having an active metal at a terminal thereof with a compound selected from the group consisting of: N-substituted cyclic amides, N-substituted cyclic ureas, N-substituted amino ketones, aromatic isocyanates, N,N,N',N'-tetramethylthiourea, N,N-disubstituted aminoalkylmethacrylamides, N-substituted aminoaldehydes, N-substituted carbodiimides, Schiff bases, propylene oxide, tetraglycidyl-1,3-bisaminomethylcyclohexane, epoxidized polybutadiene, vinyl compounds having a pyridyl group, bis(triethoxysilylpropyl)-tetrasulfide, bis(tributoxysilylpropyl)tetrasulfide, γ -glydoxypropyltrimethoxysilane,

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~~methyltrimethoxysilane,~~ tetramethoxysilane, tin tetrachloride, silicon tetrachloride,
triphenylmonochlorotin, triphenoxychlorosilane, methyltriphenoxysilane, and
diphenoxydichlorosilane.

18. (original) The rubber vulcanizate according to claim 17, which is a tire.

19. (new) The conjugated diene rubber composition according to claim 1, wherein said conjugated diene polymer chains in said rubber (A) and said rubber (B) comprise 50 to 100% by weight of conjugated diene monomer and 50 to 0% by weight of aromatic vinyl monomer.